

THEORETICAL MODEL TO DESCRIBE DISPERSIVE NONLINEAR PROPERTIES OF LEAD ZIRCONATE-TITANATE CERAMICS

K. Van Den Abeele* and M. A. Breazeale^o

National Center for Physical Acoustics
University of Mississippi
University, MS 38677



ABSTRACT

Frequency dependence of the first ultrasonic nonlinear parameter and the abnormally high third harmonic signals measured in lead zirconate-titanate (PZT) ceramics suggest the introduction of a revised theoretical model combining higher order nonlinearity and generalized dispersion. The new nonlinear dispersive equation has been solved by perturbation theory. We find a solution in the form of a set of parameters whose magnitude is obtained from a fit of the experimental data. The parameters are independent of frequency and initial amplitude. The model is applied to four samples, and the results are discussed. The validity of the perturbation theory in these cases is tested.

* Post-doctoral Research Fellow of the Belgian National Fund for Scientific Research and of Los Alamos National Laboratory, EES-4, Los Alamos, NM 87545, U.S.A. . Presently with a NATO grant at NCPA. (Permanent address: K.U.Leuven Campus Kortrijk, Interdisciplinary Research Center, B-8500 Kortrijk, Belgium)

^o Senior Scientist, NCPA. Also Professor of Physics, The University of Tennessee, Knoxville, TN 37996-1200, U.S.A.

I. INTRODUCTION

Peculiarities of sound wave behavior in crystals is shifting the attention of scientists from the linear theory to more complicated models which describe phenomena like dissipation, dispersion, and/or nonlinear propagation. To describe sound propagation in solids in the linear approximation (Hooke's law approximation) one can write the longitudinal wave equation in the form

$$\rho_o \frac{\partial^2 U}{\partial t^2} = M_2 \frac{\partial^2 U}{\partial a^2} \quad (1)$$

where ρ_o is the unstrained mass density, U is the longitudinal displacement, a is the distance measured along the propagation direction in the unstrained crystal and M_2 is a linear combination of second order elastic constants depending on the direction of propagation ($M_2 = K_2$, with K_2 as listed in Table 1). This formulation is convenient because it allows one to account for a number of phenomena in a straightforward way. For **absorption**, one simply allows complex values of M_2 .

To describe **nonlinearity** one can account for propagation in a pure mode direction (for cubic lattices one of the three principal directions), by writing the differential wave equation in the form¹:

$$\rho_o \frac{\partial^2 U}{\partial t^2} = \frac{\partial^2 U}{\partial a^2} \left[M_2 + M_3 \frac{\partial U}{\partial a} + M_4 \left(\frac{\partial U}{\partial a} \right)^2 + \dots \right] \quad (2)$$

where $M_3 = 3K_2 + K_3$ is a combination of both second and third order elastic constants, also depending on the direction of propagation (see Table 1). M_4 contains elastic constants up to the fourth order ($M_4 = \frac{3}{2}K_2 + 3K_3 + \frac{1}{2}K_4$).

For **dispersion** one can modify Eq.(1) by inclusion of a fourth order derivative with respect to the propagation distance:

$$\rho_o \frac{\partial^2 U}{\partial t^2} = M_2 \frac{\partial^2 U}{\partial a^2} + \Gamma_2 \frac{\partial^4 U}{\partial a^4} \quad (3)$$

where Γ_2 is the dispersion constant.

The solution of Eq.(2) accounts for the generation of second harmonics (and higher harmonics) during the propagation of an initially sinusoidal wave. This solution can be obtained through use of a perturbation technique²⁻⁴ or a more complicated Fourier analysis¹. Such a solution has led to the introduction of the nonlinearity parameter, the negative ratio of the coefficient of the nonlinear term to that of the linear term in the nonlinear wave equation,

$$\beta = -\frac{3K_2 + K_3}{K_2} = \frac{8A_2}{A_1^2 k^2 a} \quad (4)$$

where A_1 and A_2 are the measured amplitudes of the fundamental and its generated second harmonic; $k=2\pi/\lambda$ is the propagation constant. If the amplitude of the initial ultrasonic wave is small enough, the amplitude of the third harmonic signal, A_3 , is expressed as¹

$$A_3 = \frac{A_1^3 a^2 k^4}{32} \left(\frac{3K_2 + K_3}{K_2} \right)^2 \sqrt{1 + \frac{16}{9k^2 a^2} \left[1 - \frac{K_2(K_4 + 6K_3 + 3K_2)}{2(K_3 + 3K_2)^2} \right]^2} \quad (5)$$

in which K_4 is a combination of fourth order elastic constants. In Cu single crystals (and almost all other crystals) the amplitude of K_4 is of the order of $10K_3$. In experimental situations using ultrasonic frequencies, $k^2 a^2$ is generally of the order of 10^5 , so that for most crystalline samples one can make the approximation

$$A_3 \cong \frac{A_1^3 a^2 k^4}{32} \left(\frac{3K_2 + K_3}{K_2} \right)^2 . \quad (6)$$

Recently Na and Breazeale⁵ found that the third harmonic measured in lead zirconate-titanate (PZT) samples was much too large to allow them to make the approximation given in Eq.(6). To satisfy their data they introduced a second nonlinearity parameter which was expressed in terms of measured quantities as:

$$\beta_2 = \frac{32}{a^2 k^4} \frac{A_3}{A_1^3} . \quad (7)$$

For most crystalline solids this would mean that

$$\beta_2 = \beta^2 ; \quad (8)$$

however, its definition allowed flexibility in data interpretation for PZT. Na and Breazeale stated that serious deviations from Eq.(8) in experimental data implies that K_4 no longer is negligible and/or that a nonlin-

ear equation different from Eq.(2) must be used to describe the nonlinear wave propagation.

For single crystals, determination of the nonlinearity parameter from velocity measurements and harmonic generation yields values for the third order elastic constants which agree with other methods⁶. The results are independent of frequency. Also, the relationship given by Eq.(8) is followed for single crystals whenever it has been tested³. This means that fourth order elastic constants in single crystals are indeed negligible.

When the nonlinear properties of PZT were investigated they were found to be considerably different from those of single crystals. Na and Breazeale used their measurements to report for the first time a frequency dependence of the nonlinearity parameter β at room temperature. In addition, they found that for their PZT samples the quantities β_2 do not satisfy Eq.(8) at 10 MHz. The observed third harmonic amplitudes were found to be much larger in PZT than one would calculate from Eq.(6).

In this paper we focus on the doubly anomalous behavior of PZT ceramics and propose a solution from theoretical analysis. The suggestions of Na and Breazeale about the role of large fourth order elastic constants and/or the use of a different nonlinear equation have served as a starting point for this theoretical investigation. First, we formulate the model by combining the nonlinear equation of Thurston and Shapiro (in which we assume that K_4 is non-negligible) with a generalization of

the dispersion equation. Then we use the perturbation method to find an approximate solution which we apply and discuss in connection with the physical properties of polarized and unpolarized K1 and S1 PZT samples (Table 2). Finally, we examine the error made by using perturbation theory in our model.

II. THEORETICAL MODEL

A. Generalization of the differential equation

The third harmonic signals observed by Na and Breazeale⁵ were too large to satisfy Eq.(8), in which the influence of K_4 is considered to be negligible. For the unpolarized K1 sample at 10 MHz they observed a value of $\beta_2 = 103.8$, whereas β^2 would be only 57.8. For the S1 polarized sample at the same frequency the ratio of β_2 to β^2 is even more strikingly different from unity: $\beta_2/\beta^2 = 127$.

Since both K_2 and K_3 are known from the measurement of the velocity and the nonlinearity parameter at low frequencies (e.g., $K_2 = 14.75 \times 10^{10} \text{ kg/ms}^2$ and $K_3 = -156 \times 10^{10} \text{ kg/ms}^2$ for an unpolarized K1-sample), we can consider Eq.(5) as a function of K_4 only. Substituting this equation into Eq.(7), we obtain an expression for β_2 as a function of the fourth order elastic constant. Knowing the experimental β_2 value, this relation can be inverted numerically for K_4 or one can estimate the fourth order elastic constant from the intersection points of the graphs in Figure 1. We have found that the experimental β_2 value for the K1

sample can only be reached for a value of K_4 which is at least three orders of magnitude larger than K_3 . In an analogous way we have found that the values of K_4 for the other PZT samples must be even larger: almost five orders of magnitude for the polarized S1 sample.

As a consequence of these large K_4 values, the quantity M_4 ($=3/2 K_2 + 3K_3 + 1/2 K_4$) in Eq.(2) must be large as well. This means that this term is the most important term in the expression for the third harmonic signal amplitude. In this situation we introduce an approximation that replaces Eq.(6):

$$A_3 \cong \frac{ak^3 A_1^3}{24} \frac{M_4}{M_2}. \quad (9)$$

Indirectly, this expression calls for a new definition of β_2 . This new definition, which is distinguished by a prime, is

$$\beta_2' = \frac{24}{ak^3} \frac{A_3}{A_1^3}. \quad (10)$$

This new definition makes it possible to obtain an approximate value of the fourth order elastic constant in cases where its influence is non-negligible. The third harmonic signal measurements of Na and Breazeale have been analyzed in this way. They suggest that higher order elastic constants should be taken into account in the nonlinear differential equation.

Consequently, to make further investigation we start with the general nonlinear differential equation given by Thurston and Shapiro¹:

$$\rho_o \frac{\partial^2 U}{\partial t^2} = \frac{\partial^2 U}{\partial a^2} g\left(\frac{\partial U}{\partial a}\right) \quad (11)$$

with

$$g\left(\frac{\partial U}{\partial a}\right) = \sum_{n=2}^{\infty} M_n \left(\frac{\partial U}{\partial a}\right)^{n-2} = M_2 + M_3 \frac{\partial U}{\partial a} + M_4 \left(\frac{\partial U}{\partial a}\right)^2 + M_5 \left(\frac{\partial U}{\partial a}\right)^3 + \dots \quad (11a)$$

In comparison with the linear equation, it is worthwhile to note that the multiplier of $\frac{\partial^2 U}{\partial a^2}$ is no longer a constant. It is a series expansion in the strain $\frac{\partial U}{\partial a}$.

The dispersion effects are included in a first approximation by modifying the linear wave equation with a term proportional to the fourth derivative of the displacement with respect to the propagation distance, in analogy with the generalization of the linear wave equation [Eq.(3)]. We have found that this still does not give an adequate nonlinear equation. Therefore, we have replaced Γ_2 in Eq.(3) by a series expansion in the strain $\frac{\partial U}{\partial a}$. The combination of both nonlinear phenomena and dispersion effects lead to the following equation:

$$\rho_o \frac{\partial^2 U}{\partial a^2} = g\left(\frac{\partial U}{\partial a}\right) \frac{\partial^2 U}{\partial a^2} + h\left(\frac{\partial U}{\partial a}\right) \frac{\partial^4 U}{\partial a^4} \quad , \quad (12)$$

where

$$g\left(\frac{\partial U}{\partial a}\right) = M_2 + M_3 \frac{\partial U}{\partial a} + M_4 \left(\frac{\partial U}{\partial a}\right)^2 + M_5 \left(\frac{\partial U}{\partial a}\right)^3 + \dots \quad (12a)$$

and

$$h\left(\frac{\partial U}{\partial a}\right) = \Gamma_2 + \Gamma_3 \left(\frac{\partial U}{\partial a}\right) + \Gamma_4 \left(\frac{\partial U}{\partial a}\right)^2 + \Gamma_5 \left(\frac{\partial U}{\partial a}\right)^3 + \dots \quad (12b)$$

The purpose of our investigation is to determine the number and magnitudes of terms required in Eq.(12) for an adequate description of the behavior of PZT.

B. Approximate solution

Even though β , and especially β_2 , can be large for PZT ceramics, the second and third harmonic amplitudes measured during the experiments are still small compared with the fundamental amplitude. This means that we are looking for small perturbations of an initially well-known waveform, so that we can use perturbation theory to find a solution to Eq.(12), and later check the validity of this approach.

We rewrite Eq.(12) in the form

$$\rho_o \frac{\partial^2 U}{\partial t^2} - M_2 \frac{\partial^2 U}{\partial a^2} - \Gamma_2 \frac{\partial^4 U}{\partial a^4} = \left[g\left(\frac{\partial U}{\partial a}\right) - M_2 \right] \frac{\partial^2 U}{\partial a^2} + \left[h\left(\frac{\partial U}{\partial a}\right) - \Gamma_2 \right] \frac{\partial^4 U}{\partial a^4} \quad (13)$$

and propose a solution of this dispersive nonlinear equation in the form

$$U = U^o + U^c \quad (14)$$

with U^o the solution of the simplest dispersive linear equation:

$$\rho_o \frac{\partial^2 U^o}{\partial t^2} - M_2 \frac{\partial^2 U^o}{\partial a^2} - \Gamma_2 \frac{\partial^4 U^o}{\partial a^4} = 0 \quad , \quad (15)$$

namely,

$$U^o = A \sin(ka - \omega t) \quad \text{with} \quad \omega = \sqrt{\frac{M_2}{\rho_o}} \, k \left(1 - \frac{\Gamma_2}{M_2} k^2 \right)^{1/2} , \quad (16)$$

in which A denotes the amplitude of the sinusoidal wave at input (zero propagation distance). Substituting Eq.(14) into Eq.(13) and taking into consideration only the largest contributions on the right hand side (the zero approximation in terms of the small factors containing U^c , $\frac{\partial U^c}{\partial a}$, $\frac{\partial^2 U^c}{\partial a^2}$, ...), we find that the correction term U^c must satisfy

$$\begin{aligned} \rho_o \frac{\partial^2 U^c}{\partial t^2} - M_2 \frac{\partial^2 U^c}{\partial a^2} - \Gamma_2 \frac{\partial^4 U^c}{\partial a^4} = & \left[M_3 \frac{\partial U^o}{\partial a} + M_4 \left(\frac{\partial U^o}{\partial a} \right)^2 + M_5 \left(\frac{\partial U^o}{\partial a} \right)^3 + \dots \right] \frac{\partial^2 U^o}{\partial a^2} \\ & + \left[\Gamma_3 \frac{\partial U^o}{\partial a} + \Gamma_4 \left(\frac{\partial U^o}{\partial a} \right)^2 + \Gamma_5 \left(\frac{\partial U^o}{\partial a} \right)^3 + \dots \right] \frac{\partial U^o}{\partial a^4} . \end{aligned} \quad (17)$$

Substituting the zero approximation solution U^o into Eq.(17), this can be written in the form

$$\rho_o \frac{\partial^2 U^c}{\partial t^2} - M_2 \frac{\partial^2 U^c}{\partial a^2} - \Gamma_2 \frac{\partial^4 U^c}{\partial a^4} = \sum_{n=1}^{\infty} X_n \sin[n(ka - \omega t)] \quad (18)$$

where the X_n are:

$$X_1 = -\sum_{\ell=1}^{\infty} \frac{(M_{2\ell+2} - k^2 \Gamma_{2\ell+2}) k^{2\ell+2} A^{2\ell+1}}{2^{2\ell}} \frac{1}{2\ell+1} \binom{2\ell+1}{\ell} \quad (18a)$$

$$X_{2j} = -\sum_{\ell=j}^{\infty} \frac{(M_{2\ell+1} - k^2 \Gamma_{2\ell+1}) k^{2\ell+1} A^{2\ell}}{2^{2\ell-1}} \frac{j}{\ell} \binom{2\ell}{\ell-j} \quad (18b)$$

$$X_{2j+1} = -\sum_{\ell=j}^{\infty} \frac{(M_{2\ell+2} - k^2 \Gamma_{2\ell+2}) k^{2\ell+2} A^{2\ell+1}}{2^{2\ell}} \frac{2j+1}{2\ell+1} \binom{2\ell+1}{\ell-j}. \quad (18c)$$

In Eqs.(18a), (18b), and (18c), the final factors in each term are binomial coefficients defined as follows:

$$\binom{n}{m} = \frac{n!}{m!(n-m)!}. \quad (19)$$

For example, if one considers only the coefficients M_n and Γ_n with $n \leq 6$, Eq.(18) becomes

$$\begin{aligned} \rho_o \frac{\partial^2 U^c}{\partial t^2} - M_2 \frac{\partial^2 U^c}{\partial a^2} - \Gamma_2 \frac{\partial^4 U^c}{\partial a^4} = & - \left[\frac{(M_4 - k^2 \Gamma_4) k^4 A^3}{4} + \frac{(M_6 - k^2 \Gamma_6) k^6 A^5}{8} \right] \sin[ka - \omega t] \\ & - \left[\frac{(M_3 - k^2 \Gamma_3) k^3 A^2}{2} + \frac{(M_5 - k^2 \Gamma_5) k^5 A^4}{4} \right] \sin[2(ka - \omega t)] \\ & - \left[\frac{(M_4 - k^2 \Gamma_4) k^4 A^3}{4} + \frac{3(M_6 - k^2 \Gamma_6) k^6 A^5}{16} \right] \sin[3(ka - \omega t)] \\ & - \left[\frac{(M_5 - k^2 \Gamma_5) k^5 A^4}{8} \right] \sin[4(ka - \omega t)] \\ & - \left[\frac{(M_6 - k^2 \Gamma_6) k^6 A^5}{16} \right] \sin[5(ka - \omega t)] \end{aligned} \quad (20)$$

From Eq.(18b) one notices that the coefficients X_n for n even are influenced only by the nonlinear coefficients M_ℓ and dispersive constants Γ_ℓ having odd indices. Similarly Eqs.(18a and c) show that X_n for n odd is affected by nonlinear and dispersive coefficients having even indices.

In acoustics dispersion usually is negligible. Thus, Γ_2 is very small. If Γ_2 were identically zero, the exact solution of Eq.(18) would be

$$U^c = \sum_{n=1}^{\infty} \frac{aX_n}{2nkM_2} \cos[n(ka - \omega t)] \quad (21)$$

Let us now assume that Γ_2 is very small, but not zero. In this case, we introduce a more general series expansion

$$U^c = \sum_{n=1}^{\infty} aB_n \sin[n(ka - \omega t)] + aC_n \cos[n(ka - \omega t)] \quad (22)$$

as a solution of Eq.(18). The coefficients B_n and C_n can be dependent on the propagation distance a , but we will assume that their derivatives with respect to distance is negligible. By using this substitution and approximation, we find closed expressions for the coefficients B_n and C_n :

$$B_n = \frac{-n^2(n^2 - 1)a\Gamma_2 k^4 X_n}{n^4(n^2 - 1)^2 a^2 \Gamma_2^2 k^8 + 4k^2 n^2 (M_2 - 2n^2 \Gamma_2 k^2)^2} \quad (23a)$$

$$C_n = \frac{2nk(M_2 - 2n^2 \Gamma_2 k^2)X_n}{n^4(n^2 - 1)^2 a^2 \Gamma_2^2 k^8 + 4k^2 n^2 (M_2 - 2n^2 \Gamma_2 k^2)^2}. \quad (23b)$$

Using these expressions, one can write the amplitude A_n of the n^{th} harmonic signal:

$$A_n = \frac{a |X_n|}{\left[n^4 (n^2 - 1)^2 a^2 \Gamma_2^2 k^8 + 4k^2 n^2 (M_2 - 2n^2 \Gamma_2 k^2)^2 \right]^{1/2}} \quad (24)$$

The amplitudes of the second and third harmonics generated by propagation of an initially sinusoidal wave over a distance a in a dispersive nonlinear medium can be evaluated from Eq.(24) by using $n=2$ or $n=3$ as follows:

$$A_2 = \frac{a |M_3 - k^2 \Gamma_3| k^2 A^2}{8} \frac{\left| 1 + k^2 A^2 \frac{(M_5 - k^2 \Gamma_5)}{2(M_3 - k^2 \Gamma_3)} + \dots \right|}{\left[(M_2 - 8\Gamma_2 k^2)^2 + 9a^2 \Gamma_2^2 k^6 \right]^{1/2}} \quad (25)$$

$$A_3 = \frac{a |M_4 - k^2 \Gamma_4| k^3 A^3}{24} \frac{\left| 1 + k^2 A^2 \frac{3(M_6 - k^2 \Gamma_6)}{4(M_4 - k^2 \Gamma_4)} + \dots \right|}{\left[(M_2 - 18\Gamma_2 k^2)^2 + 144a^2 \Gamma_2^2 k^6 \right]^{1/2}} . \quad (26)$$

Note that in the non-dispersive case (when all Γ_n 's are negligible) and when only M_2 , M_3 , and M_4 are to be taken into account, Eq.(25) reduces to

$$A_2 = \frac{a |M_3| k^2 A^2}{8M_2} \quad (27)$$

which agrees with Eq.(4) used to define the nonlinearity parameter. Under the same conditions the third harmonic simplifies to

$$A_3 = \frac{d|M_4|k^3 A^3}{24M_2} \quad (28)$$

which is the limit of Eq.(5) for the third harmonic amplitude given by Thurston and Shapiro for large values of the fourth order elastic constant K_4 .

The higher harmonics can be calculated in an analogous way. The presence of the factor $ak^n A^n$ in the leading term of the expression for the n^{th} harmonic means that the amplitudes of the harmonics decrease rapidly as n increases.

III. DISCUSSION

A. APPLICATION TO PZT CERAMIC SAMPLES

Now that we have obtained an analytical solution for the dispersive nonlinear differential equation in terms of nonlinear constants M_n and dispersion constants Γ_n , we can adjust the numbers and find a set of theoretical parameters to match the experimental observations. The samples under consideration are K1 and S1 samples in both polarized and unpolarized form. The velocity, density and thickness are summarized in Table 2. The experiments of Na and Breazeale have been performed at 4 different frequencies of an initially sinusoidal ultrasonic wave: 5, 10, 15, and 30 MHz. For each sample, the range of initial

amplitudes used at these frequencies is listed in Table 3. The mean value is written between brackets. We note that the applied amplitude diminishes drastically when higher frequencies are used. Experimental measurements of the second harmonic signal at the four frequencies used and application of Eq.(4) lead to the discrete values of the nonlinearity parameter β listed in Figure 2. The nonlinearity parameter shows a frequency dependent behavior. Using the solution derived in the previous paragraphs one can find a set of parameters per sample that fit each experimental data point. The values of these parameters are given in Table 4. For each sample these nonlinearity coefficients M_n and dispersion constants Γ_n are independent of applied frequency and amplitude. The nonlinearity parameter β becomes frequency dependent because of a non-zero magnitude of Γ_3 . At a specific frequency β remains independent of the input amplitude. This can be seen in Table 5 which lists the results of the theoretical model within the amplitude range of the experiment for the K1 unpolarized sample. The value of K_4 given in Table 4 was necessary for the theoretical model to produce third harmonics as large as actually observed in the experiments. The definition of β'_2 [Eq.(10)] instead of β_2 [Eq.(7)] guarantees that the theoretical value of the new second nonlinearity parameter is independent of frequency if Γ_4 is negligible. We also observe that the magnitudes of the first and this second nonlinearity parameter do not change significantly for values of K_5 between zero and 10^{18} .

The parameter sets in Table 4 were used to make a theoretical calculation of the nonlinearity parameters for all four of the samples in the frequency range between 1 and 40 MHz. The results, using interpolation

and extrapolation on the initial mean amplitudes, are shown in Figure 2 as full lines. These theoretical curves fit the experimental data points with amazingly good agreement. The results for the K1-unpolarized sample are listed in detail in Table 6. It is necessary to allow both positive and negative values of Γ_3 in the model in order to match the experimental measurements for K1 and S1 samples respectively. The link to a physical phenomenon to explain this behavior is not yet clear.

Use of this model makes possible to calculate all constants (both nonlinear and dispersive) from experimental measurements: K_2 from velocity measurements; K_3 and K_4 from the first and second nonlinearity parameters β and β'_2 at low frequencies; Γ_2 from the velocity dispersion; Γ_3 from the dispersion (frequency dependence) of the first nonlinearity parameter; Γ_4 from the dispersion of the second nonlinearity parameter, etc. Since at present there have been no measurement of the third harmonic signal at different frequencies, we have put a question mark at the position of the Γ_4 value.

B. Estimation of perturbation theory error

Use of perturbation theory always suggests that a number of terms are neglected and that only an approximate solution is found for the general problem. Therefore it is necessary to check whether the solution is being used within the range of applicability of the perturbation theory, and the magnitude of the approximation involved.

First, we can check the magnitude of the calculated amplitudes compared with the initial amplitude of the pure sinusoidal wave at input. Tables 7 and 8, calculated with the set of parameters listed in Table 4, and with K_5 equal to 10^{18} , shows that the fundamental amplitude does not change significantly within the frequency range 1-40 MHz at the interpolated mean input amplitudes. The generated amplitudes of the second and third harmonic signals appear to be measurable, and they are indeed considerably smaller than the fundamental amplitude; e.g., of the order of 2×10^{-3} for K1-unpolarized samples and 10^{-2} for S1-polarized samples at 30 MHz for the second harmonic. The higher orders have amplitudes which diminish uniformly for all frequencies.

A second check consists of investigating the error involved when we took into account only the zero approximation of the small factors containing U^c , $\frac{\partial U^c}{\partial a}$, $\frac{\partial^2 U^c}{\partial a^2}$, etc. as contributions to the right side of Eq.(13) after substitution of Eq.(14); i.e., instead of taking into account the complete right side

$$\left[g \left(\frac{\partial U}{\partial a} \right) - M_2 \right] \frac{\partial^2 U}{\partial a^2} + \left[h \left(\frac{\partial U}{\partial a} \right) - \Gamma_2 \right] \frac{\partial^4 U}{\partial a^4} \quad (29)$$

we considered only the first terms:

$$\sum_{j=1}^{\infty} M_{j+2} \left(\frac{\partial U^o}{\partial a} \right)^j \frac{\partial^2 U^o}{\partial a^2} + \Gamma_{j+2} \left(\frac{\partial U^o}{\partial a} \right)^j \frac{\partial^4 U^o}{\partial a^4} \quad \left(= \sum_{n=1}^{\infty} X_n \sin[n(ka - \omega t)] \right) \quad (30)$$

and assumed that the difference between the two is negligible.

The use of symbolic software enables us to estimate this difference. Table 9 gives the percentage of relative error introduced by the truncation. We defined

$$\text{Estimated Error (\%)} = 100 \cdot \frac{\text{Max}_1}{\text{Max}_2} \quad (31)$$

where

$$\text{Max}_1 = \text{Max}_{\text{period}} \left| \left[g \left(\frac{\partial U}{\partial a} \right) - M_2 \right] \frac{\partial^2 U}{\partial a^2} + \left[h \left(\frac{\partial U}{\partial a} \right) - \Gamma_2 \right] \frac{\partial^4 U}{\partial a^4} - \sum_{n=1}^{\infty} X_n \sin[n(ka - \omega t)] \right|$$

with $U = U^o + U^c$

$$\text{and } \text{Max}_2 = \text{Max}_{\text{period}} \left| \sum_{n=1}^{\infty} X_n \sin[n(ka - \omega t)] \right|.$$

We note that the error never exceeds 5%, except for S1 polarized samples at 30 MHz. Looking again at Table 8, we observe that the second and third harmonic amplitudes for the S1-polarized samples are indeed substantial and that it might be inaccurate to apply the perturbation theory for higher frequencies. For the other samples we may conclude that the use of the perturbation theory is justified.

IV. CONCLUSION

We propose a theoretical model which combines higher order nonlinearity and generalized dispersion effects to interpret the results of experiments on PZT ceramics reported by Na and Breazeale. The new dispersive nonlinear differential equation has been solved by perturbation theory. It provides an analytical expression for the harmonic amplitudes generated during propagation in the samples. We applied the model to K1 and S1 samples, both polarized and unpolarized, and found that the analytical solution can be fit to experimental data by means of one set of parameters in each case. The introduction of Γ_3 accounts for the measured frequency dependence of the first nonlinear parameter β . The abnormally high third harmonic signals can be explained by assuming values for fourth order elastic constants. It is important to note that the set of parameters used in the model is independent of frequency and initial amplitude. Even though the physics behind the new differential equation and the real identity of the dispersive and nonlinear constants is not completely known at the moment, it is remarkable that this generalized dispersive-nonlinear model leads to such an extremely good fit of the data. The value of K_4 was arrived at under the assumption that the dispersion term does not contribute to the magnitude of the fourth order elastic constant. We are investigating the validity of this assumption. Finally, we believe that the use of perturbation theory in these cases is justified since the generated amplitudes are small and because the relative error introduced by truncating the right hand side of the differential equation generally is less than 5%.

ACKNOWLEDGMENTS

This work was supported by NATO (Dr. Koen Van Den Abeele) and by Murata Inc., Japan (Dr. M. A. Breazeale). Dr. Koen Van Den Abeele expresses his thanks to the Research Council of the Catholic University of Leuven, Belgium and to the Belgian National Fund for Scientific Research for additional support during the NATO-grant.

FIGURE CAPTIONS

Figure 1: The influence of K_4 on the second nonlinearity parameter β_2 [Eqs.(7) and (5)] for different samples of PZT. The intersection with the horizontal line (experimental value of β_2) gives an indication of the magnitude of the fourth order elastic constant.

- a: K1-unpolarized $\rightarrow K_4 \cong 2.5 \times 10^{15} \text{ kg/ms}^2$;
- b: K1-polarized $\rightarrow K_4 \cong 2.3 \times 10^{15} \text{ kg/ms}^2$;
- c: S1-unpolarized $\rightarrow K_4 \cong 25 \times 10^{15} \text{ kg/ms}^2$;
- d: S1-polarized $\rightarrow K_4 \cong 75 \times 10^{15} \text{ kg/ms}^2$;

Figure 2: Frequency dependence of the nonlinearity parameter β for different PZT samples. Data points represent experimental measurements. The lines are the theoretical prediction using the perturbation solution of the dispersive nonlinear differential equation with parameter values given in Table 4.

TABLE CAPTIONS

Table 1: K_2 and K_3 for [100], [110], and [111] directions.

Table 2: Physical dimensions and properties of the K1 and S1 samples of PZT ceramic.

Table 3: Range of amplitudes (10^{-10} m) used in the experiments of Na.

Table 4: List of elastic constants (kg/ms^2) and dispersion constants ($\text{kg m}/\text{s}^2$) for K1 and S1 samples.

Table 5: First and second nonlinearity parameter of the unpolarized K1 sample calculated by the dispersive nonlinear model for different amplitudes at 5, 10, 15, and 30 MHz.

Table 6: First nonlinearity parameter of the unpolarized K1 sample calculated by the dispersive nonlinear model in the frequency range 1 to 40 MHz.

Table 7: Calculated relative amplitudes of second to seventh harmonics resulting from propagation over 9.03 mm in the K1-unpolarized sample. Amplitude of fundamental at input is given as well as its relative change at the receiver position; K_2 , K_3 , K_4 , Γ_2 and Γ_3 as in Table 4; we assume $K_5 = 10^{18}$; $K_6, K_7, \dots = \Gamma_4, \Gamma_5, \dots = 0$.

Table 8: Same as Table 7, for a propagation distance of 8.82 mm in the S1-polarized sample.

Table 9: Estimated difference (%) between right hand side of the complete dispersive nonlinear differential equation and the part considered using the perturbation method.

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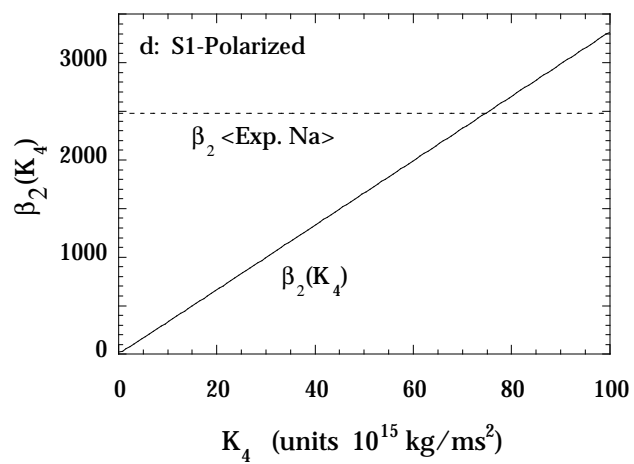
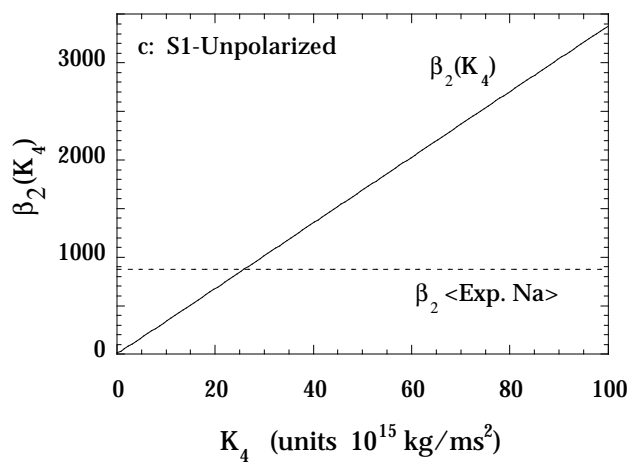
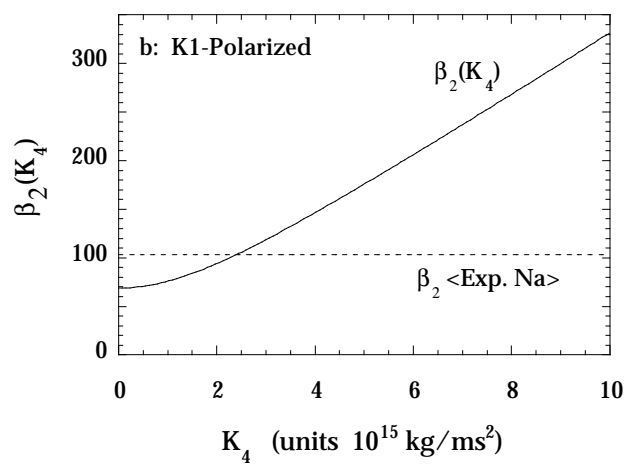
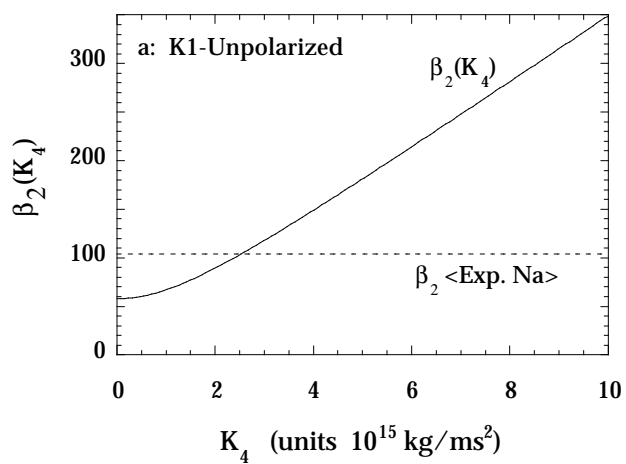


Figure 1

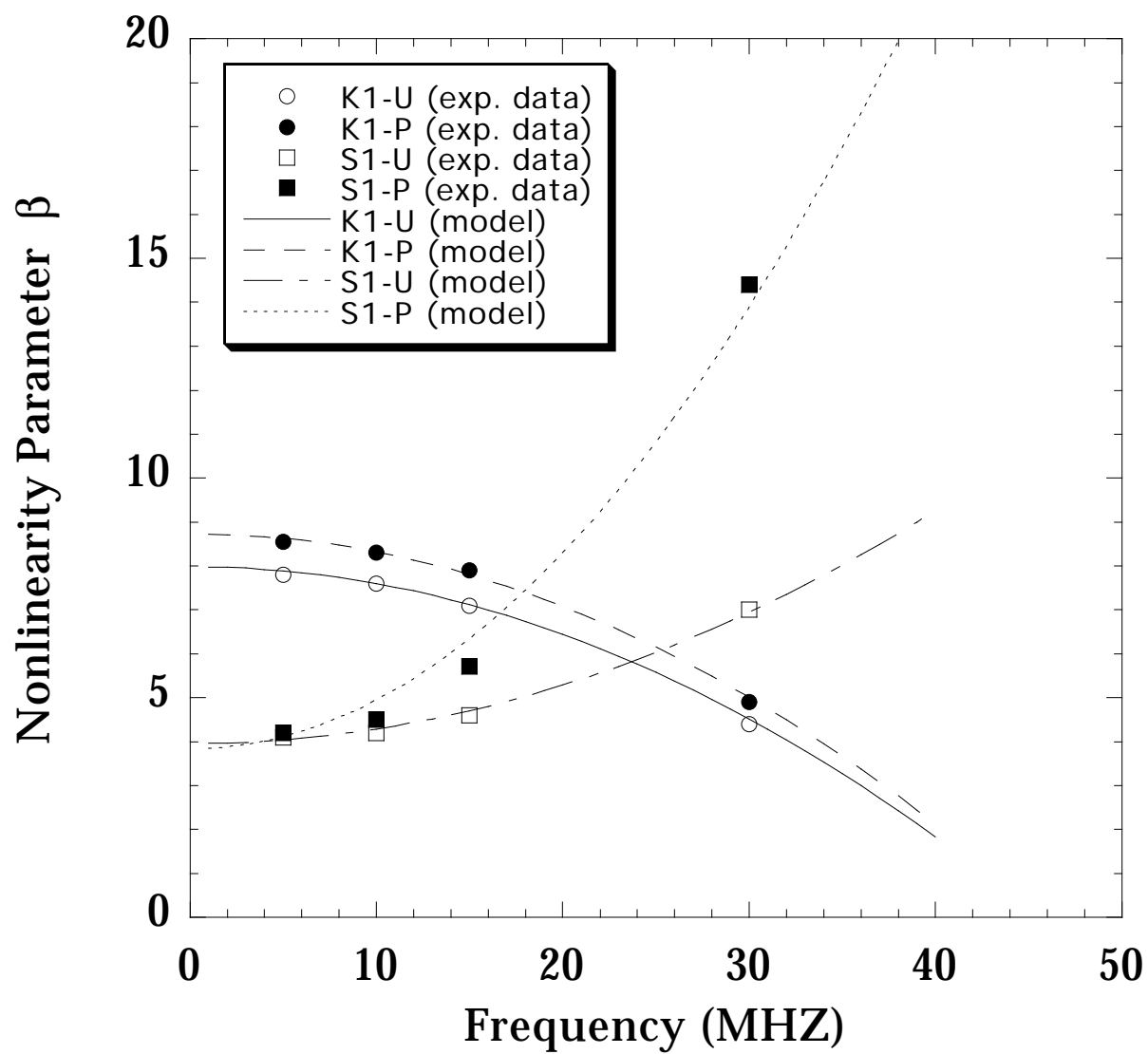


FIGURE 2

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Table 1:

Direction	K_2	K_3
[100]	C_{11}	C_{111}
[110]	$\frac{C_{11} + C_{12} + 2C_{44}}{2}$	$\frac{C_{111} + C_{112} + 2C_{166}}{4}$
[111]	$\frac{C_{11} + 2C_{12} + 4C_{44}}{3}$	$\frac{C_{111} + 6C_{112} + 12C_{144} + 24C_{166} + 2C_{123} + 16C_{456}}{9}$

Table 2:

Sample	Velocity	Density	Thickness
K1-Unpolarised	4334.1 m/s	7850 kg/m ³	9.03 ·10 ⁻³ m
K1-Polarised	4577.2 m/s	7850 kg/m ³	9.07 ·10 ⁻³ m
S1-Unpolarised	4320.0 m/s	8010 kg/m ³	9.07 ·10 ⁻³ m
S1-Polarised	4523.1 m/s	8010 kg/m ³	8.82 ·10 ⁻³ m

Table 3:

Frequency	K1-Unpolarised	K1-Polarised	S1-Unpolarised	S1-Polarised
5 MHz	16.1 - 26.2 (21.15)	18 - 27.8 (22.9)	15.8 - 29.4 (22.6)	17.1 - 28.6 (22.85)
10 MHz	13.7 - 22.6 (18.15)	16.1 - 23.3 (19.7)	14.2 - 28.2 (21.2)	16.6 - 27.5 (22.05)
15 MHz	4.5 - 11.7 (8.1)	5.4 - 11.4 (8.4)	5.1 - 11.6 (8.35)	6.0 - 11.2 (8.6)
30 MHz	1.9 - 3.0 (2.45)	2.2 - 3.4 (2.8)	2.0 - 4.4 (3.2)	2.1 - 4.7 (3.4)

Table 4:

Sample	K_2	K_3	K_4	G_2	G_3	G_4
K1-Unpolarised	$14.75 \cdot 10^{10}$	$-162.0 \cdot 10^{10}$	$3.015 \cdot 10^{15}$	$2.0 \cdot 10^{-3}$	$-2.70 \cdot 10^2$?
K1-Polarised	$16.45 \cdot 10^{10}$	$-193.0 \cdot 10^{10}$	$3.190 \cdot 10^{15}$	$2.0 \cdot 10^{-3}$	$-3.60 \cdot 10^2$?
S1-Unpolarised	$14.95 \cdot 10^{10}$	$-104.0 \cdot 10^{10}$	$25.830 \cdot 10^{15}$	$2.0 \cdot 10^{-3}$	$2.35 \cdot 10^2$?
S1-Polarised	$16.39 \cdot 10^{10}$	$-112.0 \cdot 10^{10}$	$74.700 \cdot 10^{15}$	$2.0 \cdot 10^{-3}$	$9.50 \cdot 10^2$?

Table 5:

Frequency (MHz)	Amplitude (10^{-10} m)	b	β'_2
5.0000	16.1000	7.8868665	10189.0192944
5.0000	17.1000	7.8868564	10189.0193498
5.0000	18.1000	7.8868456	10189.0194085
5.0000	19.1000	7.8868342	10189.0194704
5.0000	20.1000	7.8868222	10189.0195355
5.0000	21.1000	7.8868096	10189.0196039
5.0000	22.1000	7.8867964	10189.0196755
5.0000	23.1000	7.8867826	10189.0197503
5.0000	24.1000	7.8867682	10189.0198282
5.0000	25.1000	7.8867531	10189.0199094
5.0000	26.1000	7.8867375	10189.0199936
10.0000	13.7000	7.5983899	10189.3115224
10.0000	14.7000	7.5983551	10189.3116923
10.0000	15.7000	7.5983179	10189.3118707
10.0000	16.7000	7.5982783	10189.3120571
10.0000	17.7000	7.5982362	10189.3122507
10.0000	18.7000	7.5981917	10189.3124507
10.0000	19.7000	7.5981447	10189.3126563
10.0000	20.7000	7.5980953	10189.3128667
10.0000	21.7000	7.5980434	10189.3130808
15.0000	4.5000	7.1179597	10188.9037500
15.0000	5.5000	7.1179322	10188.9038908
15.0000	6.5000	7.1178992	10188.9040541
15.0000	7.5000	7.1178607	10188.9042369
15.0000	8.5000	7.1178166	10188.9044355
15.0000	9.5000	7.1177671	10188.9046459
15.0000	10.5000	7.1177120	10188.9048635
15.0000	11.5000	7.1176514	10188.9050830
30.0000	1.9000	4.5204777	10119.9357741
30.0000	2.0000	4.5204734	10119.9357936
30.0000	2.1000	4.5204689	10119.9358136
30.0000	2.2000	4.5204642	10119.9358341
30.0000	2.3000	4.5204592	10119.9358550
30.0000	2.4000	4.5204540	10119.9358762
30.0000	2.5000	4.5204486	10119.9358978
30.0000	2.6000	4.5204430	10119.9359195
30.0000	2.7000	4.5204372	10119.9359413
30.0000	2.8000	4.5204311	10119.9359631
30.0000	2.9000	4.5204249	10119.9359849
30.0000	3.0000	4.5204184	10119.9360064

Table 6:

Frequency (MHz)	Amplitude (10⁻¹⁰ m)	b
1.0000	22.2000	7.9792006
2.0000	21.9375	7.9676506
3.0000	21.6750	7.9484015
4.0000	21.4125	7.9214540
5.0000	21.1500	7.8868090
6.0000	20.5500	7.8444731
7.0000	19.9500	7.7944449
8.0000	19.3500	7.7367255
9.0000	18.7500	7.6713158
10.0000	18.1500	7.5982165
11.0000	16.1400	7.5174985
12.0000	14.1300	7.4291053
13.0000	12.1200	7.3330337
14.0000	10.1100	7.2292790
15.0000	8.1000	7.1178349
16.0000	7.7233	6.9986233
17.0000	7.3467	6.8717153
18.0000	6.9700	6.7371089
19.0000	6.5933	6.5948018
20.0000	6.2167	6.4447918
21.0000	5.8400	6.2870760
22.0000	5.4633	6.1216518
23.0000	5.0867	5.9485163
24.0000	4.7100	5.7676668
25.0000	4.3333	5.5791006
26.0000	3.9567	5.3828152
27.0000	3.5800	5.1788086
28.0000	3.2033	4.9670794
29.0000	2.8267	4.7476268
30.0000	2.4500	4.5204514
31.0000	2.3550	4.2855400
32.0000	2.2600	4.0429119
33.0000	2.1650	3.7925730
34.0000	2.0700	3.5345314
35.0000	1.9750	3.2687980
36.0000	1.8800	2.9953872
37.0000	1.7850	2.7143169
38.0000	1.6900	2.4256098
39.0000	1.5950	2.1292932
40.0000	1.5000	1.8254007

Table 7:

Frequency (MHz)	Amplitude A (10E-10 m)	A ₁ /A-1.0	A ₂ /A	A ₃ /A	A ₄ /A	A ₅ /A	A ₆ /A	A ₇ /A
1.0	22.2000	0.149E-13	0.420E-04	0.575E-07	0.793E-11	0.375E-16	0.700E-22	0.642E-28
2.0	21.9375	0.909E-12	0.166E-03	0.449E-06	0.122E-09	0.114E-14	0.422E-20	0.765E-26
3.0	21.6750	0.987E-11	0.368E-03	0.148E-05	0.598E-09	0.828E-14	0.453E-19	0.122E-24
4.0	21.4125	0.528E-10	0.644E-03	0.343E-05	0.182E-08	0.332E-13	0.239E-18	0.847E-24
5.0	21.1500	0.192E-09	0.989E-03	0.653E-05	0.429E-08	0.966E-13	0.858E-18	0.375E-23
6.0	20.5500	0.510E-09	0.138E-02	0.106E-04	0.815E-08	0.214E-12	0.222E-17	0.113E-22
7.0	19.9500	0.114E-08	0.181E-02	0.159E-04	0.138E-07	0.411E-12	0.483E-17	0.278E-22
8.0	19.3500	0.225E-08	0.227E-02	0.224E-04	0.215E-07	0.709E-12	0.923E-17	0.590E-22
9.0	18.7500	0.403E-08	0.276E-02	0.299E-04	0.314E-07	0.113E-11	0.160E-16	0.111E-21
10.0	18.1500	0.666E-08	0.327E-02	0.385E-04	0.433E-07	0.168E-11	0.255E-16	0.191E-21
11.0	16.1400	0.737E-08	0.348E-02	0.405E-04	0.446E-07	0.169E-11	0.252E-16	0.184E-21
12.0	14.1300	0.730E-08	0.358E-02	0.403E-04	0.424E-07	0.153E-11	0.218E-16	0.152E-21
13.0	12.1200	0.639E-08	0.356E-02	0.377E-04	0.369E-07	0.124E-11	0.163E-16	0.106E-21
14.0	10.1100	0.483E-08	0.340E-02	0.327E-04	0.288E-07	0.866E-12	0.103E-16	0.595E-22
15.0	8.1000	0.301E-08	0.308E-02	0.259E-04	0.195E-07	0.503E-12	0.511E-17	0.253E-22
16.0	7.7233	0.366E-08	0.328E-02	0.285E-04	0.219E-07	0.574E-12	0.591E-17	0.296E-22
17.0	7.3467	0.431E-08	0.346E-02	0.310E-04	0.240E-07	0.635E-12	0.659E-17	0.331E-22
18.0	6.9700	0.492E-08	0.361E-02	0.331E-04	0.257E-07	0.683E-12	0.708E-17	0.354E-22
19.0	6.5933	0.545E-08	0.372E-02	0.348E-04	0.270E-07	0.715E-12	0.735E-17	0.362E-22
20.0	6.2167	0.586E-08	0.380E-02	0.361E-04	0.278E-07	0.727E-12	0.736E-17	0.355E-22
21.0	5.8400	0.612E-08	0.384E-02	0.369E-04	0.279E-07	0.719E-12	0.711E-17	0.332E-22
22.0	5.4633	0.620E-08	0.384E-02	0.371E-04	0.275E-07	0.690E-12	0.662E-17	0.296E-22
23.0	5.0867	0.608E-08	0.380E-02	0.367E-04	0.265E-07	0.642E-12	0.591E-17	0.252E-22
24.0	4.7100	0.577E-08	0.371E-02	0.357E-04	0.249E-07	0.578E-12	0.506E-17	0.203E-22
25.0	4.3333	0.528E-08	0.358E-02	0.342E-04	0.227E-07	0.502E-12	0.413E-17	0.155E-22
26.0	3.9567	0.464E-08	0.341E-02	0.320E-04	0.201E-07	0.419E-12	0.321E-17	0.111E-22
27.0	3.5800	0.390E-08	0.321E-02	0.293E-04	0.173E-07	0.333E-12	0.234E-17	0.744E-23
28.0	3.2033	0.311E-08	0.296E-02	0.262E-04	0.142E-07	0.251E-12	0.160E-17	0.460E-23
29.0	2.8267	0.233E-08	0.268E-02	0.226E-04	0.112E-07	0.178E-12	0.101E-17	0.259E-23
30.0	2.4500	0.161E-08	0.236E-02	0.188E-04	0.827E-08	0.116E-12	0.574E-18	0.129E-23
31.0	2.3550	0.167E-08	0.230E-02	0.191E-04	0.830E-08	0.113E-12	0.543E-18	0.119E-23
32.0	2.2600	0.172E-08	0.222E-02	0.193E-04	0.825E-08	0.109E-12	0.505E-18	0.108E-23
33.0	2.1650	0.174E-08	0.212E-02	0.194E-04	0.810E-08	0.104E-12	0.462E-18	0.956E-24
34.0	2.0700	0.174E-08	0.201E-02	0.194E-04	0.787E-08	0.968E-13	0.415E-18	0.834E-24
35.0	1.9750	0.172E-08	0.188E-02	0.192E-04	0.756E-08	0.891E-13	0.367E-18	0.715E-24
36.0	1.8800	0.167E-08	0.173E-02	0.189E-04	0.718E-08	0.808E-13	0.319E-18	0.601E-24
37.0	1.7850	0.160E-08	0.157E-02	0.184E-04	0.673E-08	0.720E-13	0.272E-18	0.495E-24
38.0	1.6900	0.151E-08	0.140E-02	0.178E-04	0.622E-08	0.631E-13	0.227E-18	0.399E-24
39.0	1.5950	0.140E-08	0.123E-02	0.170E-04	0.568E-08	0.543E-13	0.187E-18	0.315E-24
40.0	1.5000	0.127E-08	0.104E-02	0.162E-04	0.510E-08	0.459E-13	0.150E-18	0.242E-24

Table 8:

Frequency (MHz)	Amplitude A (10E-10 m)	A ₁ /A - 1.0	A ₂ /A	A ₃ /A	A ₄ /A	A ₅ /A	A ₆ /A	A ₇ /A
1.0	23.4000	0.679E-11	0.191E-04	0.123E-05	0.117E-10	0.399E-16	0.664E-22	0.584E-28
2.0	23.2625	0.425E-09	0.768E-04	0.972E-05	0.184E-09	0.125E-14	0.413E-20	0.722E-26
3.0	23.1250	0.472E-08	0.174E-03	0.324E-04	0.913E-09	0.925E-14	0.456E-19	0.119E-24
4.0	22.9875	0.259E-07	0.314E-03	0.759E-04	0.283E-08	0.380E-13	0.249E-18	0.861E-24
5.0	22.8500	0.965E-07	0.500E-03	0.146E-03	0.680E-08	0.113E-12	0.921E-18	0.396E-23
6.0	22.6900	0.280E-06	0.736E-03	0.250E-03	0.138E-07	0.274E-12	0.266E-17	0.136E-22
7.0	22.5300	0.687E-06	0.103E-02	0.391E-03	0.250E-07	0.576E-12	0.646E-17	0.383E-22
8.0	22.3700	0.149E-05	0.139E-02	0.575E-03	0.418E-07	0.109E-11	0.139E-16	0.935E-22
9.0	22.2100	0.293E-05	0.181E-02	0.807E-03	0.655E-07	0.191E-11	0.272E-16	0.204E-21
10.0	22.0500	0.536E-05	0.232E-02	0.109E-02	0.977E-07	0.314E-11	0.493E-16	0.409E-21
11.0	19.3600	0.564E-05	0.258E-02	0.112E-02	0.968E-07	0.301E-11	0.456E-16	0.365E-21
12.0	16.6700	0.523E-05	0.278E-02	0.108E-02	0.875E-07	0.256E-11	0.363E-16	0.273E-21
13.0	13.9800	0.418E-05	0.288E-02	0.964E-03	0.711E-07	0.189E-11	0.243E-16	0.166E-21
14.0	11.2900	0.277E-05	0.284E-02	0.785E-03	0.504E-07	0.116E-11	0.130E-16	0.771E-22
15.0	8.6000	0.141E-05	0.261E-02	0.560E-03	0.293E-07	0.552E-12	0.505E-17	0.243E-22
16.0	8.2533	0.176E-05	0.301E-02	0.626E-03	0.336E-07	0.646E-12	0.604E-17	0.296E-22
17.0	7.9067	0.214E-05	0.343E-02	0.689E-03	0.376E-07	0.736E-12	0.699E-17	0.347E-22
18.0	7.5600	0.252E-05	0.389E-02	0.748E-03	0.413E-07	0.817E-12	0.783E-17	0.392E-22
19.0	7.2133	0.289E-05	0.436E-02	0.801E-03	0.445E-07	0.886E-12	0.852E-17	0.426E-22
20.0	6.8667	0.322E-05	0.485E-02	0.846E-03	0.471E-07	0.938E-12	0.899E-17	0.446E-22
21.0	6.5200	0.351E-05	0.536E-02	0.883E-03	0.490E-07	0.970E-12	0.922E-17	0.450E-22
22.0	6.1733	0.373E-05	0.588E-02	0.910E-03	0.500E-07	0.980E-12	0.916E-17	0.437E-22
23.0	5.8267	0.387E-05	0.639E-02	0.926E-03	0.502E-07	0.966E-12	0.884E-17	0.409E-22
24.0	5.4800	0.391E-05	0.690E-02	0.931E-03	0.494E-07	0.930E-12	0.825E-17	0.368E-22
25.0	5.1333	0.384E-05	0.739E-02	0.923E-03	0.477E-07	0.872E-12	0.745E-17	0.317E-22
26.0	4.7867	0.368E-05	0.784E-02	0.902E-03	0.452E-07	0.795E-12	0.649E-17	0.262E-22
27.0	4.4400	0.341E-05	0.825E-02	0.869E-03	0.418E-07	0.703E-12	0.544E-17	0.207E-22
28.0	4.0933	0.307E-05	0.860E-02	0.823E-03	0.378E-07	0.601E-12	0.437E-17	0.155E-22
29.0	3.7467	0.266E-05	0.887E-02	0.766E-03	0.332E-07	0.496E-12	0.335E-17	0.110E-22
30.0	3.4000	0.221E-05	0.905E-02	0.698E-03	0.283E-07	0.392E-12	0.243E-17	0.731E-23
31.0	3.2900	0.236E-05	0.980E-02	0.720E-03	0.290E-07	0.397E-12	0.241E-17	0.708E-23
32.0	3.1800	0.249E-05	0.106E-01	0.739E-03	0.296E-07	0.397E-12	0.235E-17	0.675E-23
33.0	3.0700	0.260E-05	0.114E-01	0.755E-03	0.299E-07	0.393E-12	0.226E-17	0.633E-23
34.0	2.9600	0.269E-05	0.122E-01	0.766E-03	0.299E-07	0.384E-12	0.214E-17	0.585E-23
35.0	2.8500	0.275E-05	0.130E-01	0.773E-03	0.297E-07	0.371E-12	0.200E-17	0.533E-23
36.0	2.7400	0.278E-05	0.138E-01	0.776E-03	0.292E-07	0.353E-12	0.184E-17	0.478E-23
37.0	2.6300	0.278E-05	0.147E-01	0.775E-03	0.285E-07	0.332E-12	0.167E-17	0.422E-23
38.0	2.5200	0.275E-05	0.155E-01	0.769E-03	0.275E-07	0.309E-12	0.149E-17	0.367E-23
39.0	2.4100	0.269E-05	0.162E-01	0.758E-03	0.263E-07	0.283E-12	0.132E-17	0.315E-23
40.0	2.3000	0.260E-05	0.170E-01	0.742E-03	0.249E-07	0.256E-12	0.114E-17	0.265E-23

Table 9:

Frequency	Amplitude	K1-Unpolarised	K1-Polarised	S1-Unpolarised	S1-Polarised
5 MHz	25 10 ⁻¹⁰ m	0.96524	0.94616	0.72935	0.98461
10 MHz	20 10 ⁻¹⁰ m	3.03273	2.96520	2.90238	4.48704
15 MHz	10 10 ⁻¹⁰ m	3.16582	3.10470	3.08937	4.79720
30 MHz	3 10 ⁻¹⁰ m	2.40786	2.39183	4.35913	8.08418